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13. ABSTRACT (Maximum 200 Words) We have purchased and installed a new tunable femtosecond (150 fs) laser system dedicated to nonlinear optical characterization of organic materials. We have conducted the first nonlinear optical measurements of a thin film endohedral metallofullerene, Gd2@C80 using the new system. Single beam z scan experiments were carried out to measure the size and the sign of the third order susceptibility at 400nm, 590nm, 648nm, 800nm, and 1000nm. At 800nm measurements were conducted for different pulse duration between 130fs and 1ps. We found that, when wavelength and pulse duration are taken into account, the third order susceptibility of Gd2@C80 is about one order of magnitude larger than that of empty-cage C60 or C70. The research has provided practical and theoretical training in high power laser technology, nonlinear optics, crystallography, and spectroscopy to three graduate and two undergraduate students				
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Organic Ultrafast Nonlinear Optical Devices Characterization of Organic Photonic Materials

Statement of the problem studied

The main goal of the proposed research was to characterize the nonlinear optical response of new organic materials and to fabricate and characterize new all optical all-optical devices. To carry out the proposed research a reliable, tunable femtosecond laser source was needed. With partial funding from this grant we purchased and installed a Ti:sapphire femtosecond laser/regenerative amplifier system consisting of a Ti:sapphire oscillator pumped by the second harmonic of a diode pumped Nd: YLF cw laser, a regenerative pumped by an Nd:YLF pulsed laser, and an optical parametric amplifier (OPA).

We used two techniques to characterize the nonlinear optical response of new materials: time resolved degenerate four wave mixing [1] and z-scan. [2] In the time resolved degenerate four wave mixing technique the femtosecond laser pulses are split with beamsplitters and directed, through variable delay lines, toward the sample. Two-time - coincident femtosecond excitation pulses and a third delayed probe beam cross in the sample. The optical interference pattern produced by the overlap of two excitation beams induces a modulation of the index of refraction of the material. In a centrosymmetric medium the modulation is proportional to the third order nonlinear coefficient. The detected signal is proportional to the absolute value of the nonlinear coefficient. The availability of a widely tunable source should allow to obtain the dispersion (linewidth and shape) of the nonlinear response. In the Z-scan approach, a single Gaussian beam is tightly focused into a thin nonlinear medium. The transmittance through a small aperture in the far field is measured. The sample is moved along the z direction in and out of focus. When the signal through the small aperture is plotted as a function of the sample position, a negative lens effect, arising from a nonlinear materials having negative n_2 , will result in a peak followed by a valley in the transmittance curve, while positive n_2 will give a curve with a valley-peak sequence as the sample is translated from -z side to +z side. Thus, the sign of the nonlinearity can be readily determined. Moreover, the size of the nonlinearity can be determined from the difference in peak and valley in transmittance.

The combination of these techniques allowed us to determine the magnitude of the real and imaginary parts and the time response of χ^3 in various materials as a function of different external parameters (wavelength, temperature).

Summary of the most important results

During the duration of this grant the following important results were obtained:

- 1) A new tunable femtosecond (150 fs) laser system has been purchased and installed (see figure 1). A four wave-mixing set-up, a zscan apparatus and a pump and probe experiment have been built and are currently in operation.
- 2) Recently, large nonlinear optical responses have been reported in solutions of endohedral metallofullerenes in which the spheroidal molecular structure is used to encapsulate atoms inside the fullerene cage. The enhancement of the nonlinear coefficient was attributed to a metal-to-cage charge-transfer

mechanism. We have conducted the first nonlinear optical measurements of a thin film endohedral metallofullerene, Gd2@C80. Single beam z scan experiments were carried out using the 130 fs tunable Ti:Sapphire-OPA laser system. The size and the sign of the third order susceptibility were measured at 400 nm, 590 nm, 648 nm, 800 nm, and 1000 nm. At 800 nm measurements were conducted for different pulse duration between 130 fs and 1 ps. Our results show that the measured values of the third order nonlinearity were relatively large ($10^{-10} < \chi^{(3)} < 10^{-9}$ esu), negative, and strongly dependent on pulse duration (longer pulses gave larger nonlinear coefficients) and wavelength (resonance enhancement at short wavelengths). We found that, when wavelength and pulse duration are taken into account, the third order susceptibility of Gd2@C80 is about one order of magnitude larger than that of empty-cage C60 or C70.

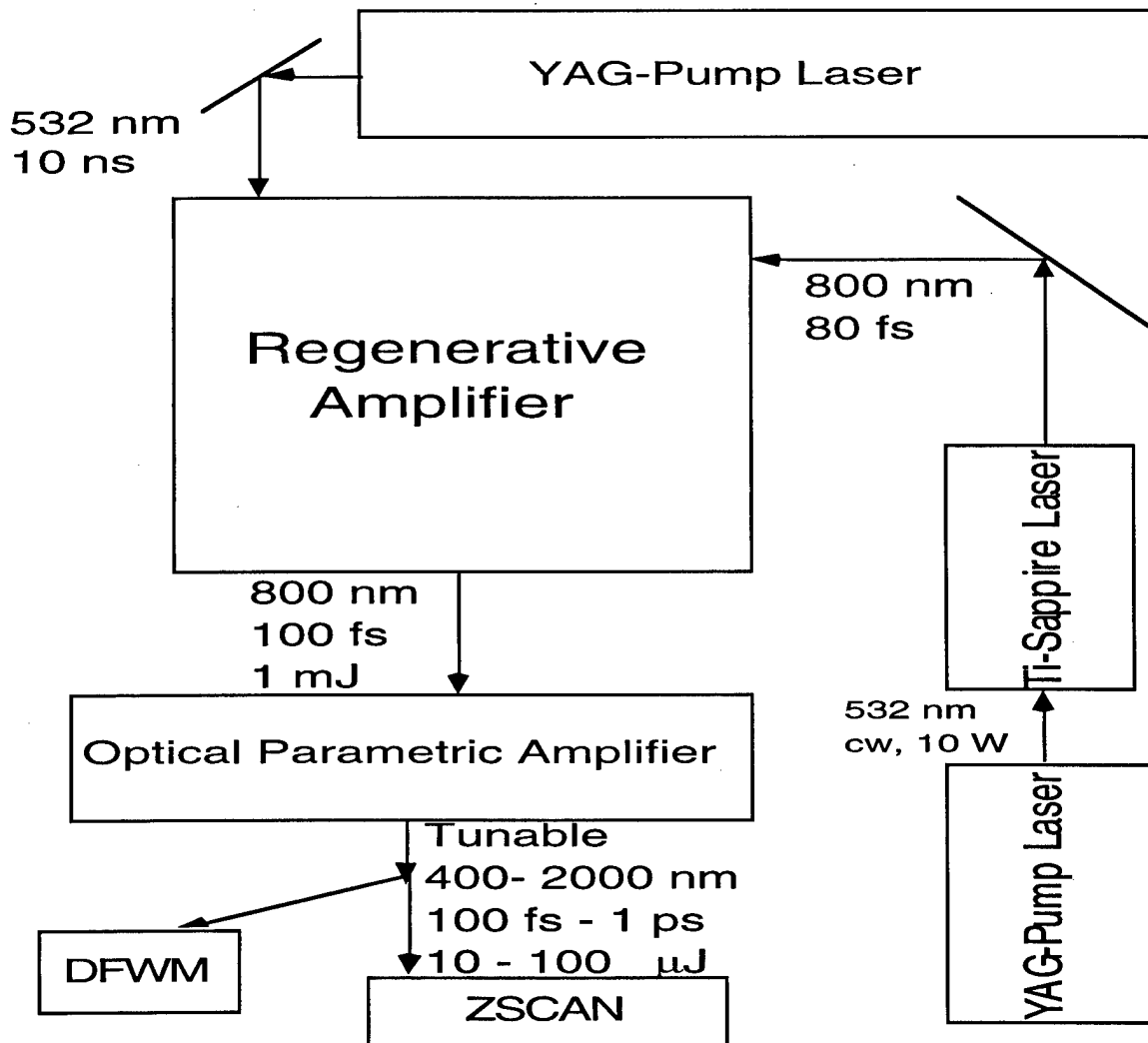


Figure 1. Femtosecond Characterization Setup

- 3) Third harmonic conversion is an effective technique to generate laser radiation at 355 nm from high power solid state lasers. Numerous studies have shown that conversion efficiency depends on parameters such as laser beam quality, crystal nonlinear coefficient, crystal optical quality and interaction geometry. Relatively little attention has been given to possible limitation in conversion efficiency due to two photon absorption at the third harmonic wavelength (355nm). Using the open aperture Z-Scan technique, we measured the two-photon coefficient beta of KDP crystals. The investigated samples were rapid grown KDP crystals doped with different impurities (i.e. Mg, Fe, Cr) at various concentration levels. The results of this investigation show that two-photon absorption is always present in KDP crystals at 355 nm and is strongly dependent on the crystal composition and morphology. The two-photon absorption coefficient beta varied from 0.001 to 0.02 cm/GW. We have observed an apparent correlation between the two-photon absorption coefficient at 355 nm and the optical density at 260 nm with respect to the distribution of the doping concentration. In addition, we confirmed that impurities seem to not only prefer to reside in the prismatic sector of the crystal (as opposed to the pyramidal), but also their distribution within the high impurity sector is not uniform.
- 4) The program has provided practical and theoretical training in high power laser technology, nonlinear optics, crystallography, and spectroscopy to three graduate and two undergraduate students. One graduate student (David Harris) has obtained a Ph.D. and is now a research scientist with a large photonics private company.

List of Publications

Nonlinear optical response of Gd₂O₃ thin films. Gul Coskun , Roger Dorsinville.
Abstract submitted to SPIE 44th Annual Meeting 18-23- July 1999 Colorado Convention Center Denver, Colorado

Nonlinear Optical absorption and refraction in Optical Crystals at 355 nm. Ardie D. Walser, Adil Bouselhami, Roger Dorsinville. Abstract submitted to SPIE 44th Annual Meeting 18-23- July 1999 Colorado Convention Center Denver, Colorado.

Participating Scientific Personnel

Prof. Roger Dorsinville (PI).

Prof. Ardie Walser

Dr. David Harris (Ph.D. December 97)

Adil Bouselhami (B. Sc. December 98)

Gul Coskun, Electrical Engineering graduate student

Robinson Pino, Electrical Engineering undergraduate student

Vivian Kweong, Electrical Engineering graduate student

Report of Inventions

N/A

Bibliography

- 1) Excited-state dynamics of thiophene thin films studied by time-resolved degenerate four-wave mixing. David Harris, Takeo Mukai, and Roger Dorsinville. Appl Phys. Lett., **70**, 1216 (1997)
- 2) Experimental Observation of Magnitude and sign of the Third Order Optical Nonlinearity in Polydibutylstannane. R. Priestley, A. D. Walser, R. Dorsinville, W.K. Zou, D.Y. Xu, and N.-L. Yang. Optics Communications, **347**, 131 (1996).